

Europäisches Patentamt

European Patent Office

Office européen des brevets



(11) EP 1 156 546 A1

(12)

EUROPEAN PATENT APPLICATION

(43) Date of publication: 21.11.2001 Bulletin 2001/47

(51) Int Cl.7: **H01M 8/24**, H01M 8/02, H01M 8/10

(21) Application number: 01119398.4

(22) Date of filing: 15.07.1998

(84) Designated Contracting States:

AT BE CH CY DE DK ES FI FR GB GR IE IT LI LU

MC NL PT SE

(30) Priority: 16.07.1997 US 52713 P

(62) Document number(s) of the earlier application(s) in accordance with Art. 76 EPC: 98933412.3 / 1 018 177

(71) Applicant: BALLARD POWER SYSTEMS INC. Burnaby, British Columbia V5J 5J9 (CA) (72) Inventor: The designation of the inventor has not yet been filed

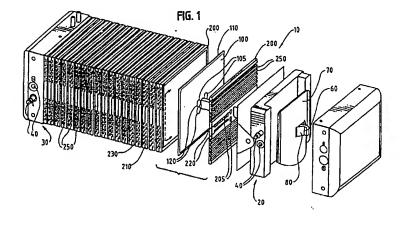
(74) Representative: Weller, Erich W. Patentanwälte Ruff, Wilhelm, Beler, Dauster & Partner Postfach 10 40 36 70035 Stuttgart (DE)

Remarks:

Diese Anmeldung ist am 11 - 08 - 2001 als Teilanmeldung zu der unter INID-Kode 62 erwähnten Anmeldung eingereicht worden.

- (54) Method of making a resilient seal for membrane electrode assembly (MEA) in an electrochemical fuel cell
- (57) An improved method of making a membrane electrode assembly ("MEA") for an electrochemical fuel cell is provided, which MEA comprises coextensive ion exchange membrane and electrode layers and a resilient fluid impermeable integral seal comprising a sealant material impregnated into the porous electrode layers in sealing regions of the MEA. The MEA is made from a multi-layer material by impregnating sealing regions with a flow processable sealant material, curing same, and cutting through said multi-layer material in said sealing regions. In preferred embodiments, the integral seal circumscribes the electrochemically active area of the

MEA. The integral seal preferably also extends laterally beyond the edge of the MEA, thereby envelopping the peripheral region including the side edge of the MEA. An integral seal may also be provided around any openings, such as manifold openings, that are formed in the MEA. Preferably the sealant material is a flow processable elastomer applied to the MEA using an injection molding process prior to being cured. In preferred embodiments the seal has a plurality of spaced parallel raised ribs with cross ribs extending therebetween at spaced intervals. The raised ribs and cross ribs provide compartmentalized seals that provide improved protection against fluid leaks in a fuel cell assembly.



Description

Technical Field

[0001] The present invention relates to electrochemical fuel cells. In particular the invention provides a method of making an improved membrane electrode assembly. An improved membrane electrode assembly comprises integral fluid impermeable and coextensive electrode and membrane layers.

Background

[0002] Electrochemical fuel cells convert reactants, namely fuel and oxidant fluid streams, to generate electric power and reaction products. Electrochemical fuel cells employ an electrolyte disposed between two electrodes, namely a cathode and an anode. The electrodes each comprise an electrocatalyst disposed at the interface between the electrolyte and the electrodes to induce the desired electrochemical reactions. The location of the electrocatalyst generally defines the electrochemically active area.

[0003] Solid polymer fuel cells generally employ a membrane electrode assembly ("MEA") consisting of a solid polymer electrolyte or ion exchange membrane disposed between two electrode layers comprising porous, electrically conductive sheet material. The membrane is ion conductive (typically proton conductive), and also acts as a barrier for isolating the reactant streams from each other. Another function of the membrane is to act as an electrical insulator between the two electrode layers. The electrodes must be electrically insulated from each other to prevent short-circuiting. If a multi-layer MEA is cut, tiny portions of the electrically conductive electrode material, such as stray fibers, may bridge across the thin membrane, interconnecting the electrodes, which could cause electrical short-circuiting in an operating fuel cell. Conventional MEAs incorporate a membrane with a larger surface area than the electrode layers, with at least a small portion of the membrane extending laterally beyond the edge of the electrode layers. The protruding membrane edge helps to prevent short-circuiting between the electrodes around the edge of the membrane. A problem with this is that it is difficult to cut an MEA after the electrodes have been joined to the membrane so that the thin membrane has a larger area than the electrodes. A conventional MEA is fabricated by manufacturing and cutting the electrodes and membrane layers separately. After the electrodes and membrane have been cut to the desired size and shape, the cut electrode layers are laminated with the cut membrane layer. These steps are not conducive to high speed manufacturing processes. It would be preferable to manufacture a sheet or roll of MEA material that already comprises the electrode and membrane layers, wherein this multi-layer material could then be cut to the desired size and shape for individual MEAs.

An MEA cut in this way, such that the electrodes and membrane are coextensive, is described herein as being a "flush cut" MEA. However, this approach has heretofore been impractical because of the short circuiting problem described above.

[0004] In a fuel cell stack, the MEA is typically interposed between two separator plates that are substantially impermeable to the reactant fluid streams. The plates act as current collectors and provide support for the electrodes. To control the distribution of the reactant fluid streams to the electrochemically active area, the surfaces of the plates that face the MEA may have openfaced channels or grooves formed therein. Such channels or grooves define a flow field area that generally corresponds to the adjacent electrochemically active area. Such separator plates, which have reactant channels formed therein are commonly known as flow field plates. In a fuel cell stack a plurality of fuel cells are connected together, typically in series, to increase the overall output power of the assembly. In such an arrangement, one side of a given plate may serve as an anode plate for one cell and the other side of the plate may serve as the cathode plate for the adjacent cell. In this arrangement the plates may be referred to as bipolar plates.

[0005] The fuel fluid stream that is supplied to the anode typically comprises hydrogen. For example, the fuel fluid stream may be a gas such as substantially pure hydrogen or a reformate stream containing hydrogen. Alternatively, a liquid fuel stream such as aqueous methanol may be used. The oxidant fluid stream, which is supplied to the cathode, typically comprises oxygen, such as substantially pure oxygen, or a dilute oxygen stream such as air. In a fuel cell stack, the reactant streams arc typically supplied and exhausted by respective supply and exhaust manifolds. Manifold ports are provided to fluidly connect the manifolds to the flow field area and electrodes. Manifolds and corresponding ports may also be provided for circulating a coolant fluid through interior passages within the stack to absorb heat generated by the exothermic fuel cell reactions.

[0006] It is desirable to seal reactant fluid stream passages to prevent leaks or intermixing of the fuel and oxidant fluid streams. Fuel cell stacks typically employ resilient seals between stack components. Such seals isolate the manifolds and the electrochemically active area of the fuel cell MEAs by circumscribing these areas. For example, a fluid tight seal may be achieved in a conventional fuel cell stack by using elastomeric gasket seals interposed between the flow field plates and the membrane, with sealing effected by applying a compressive force to the resillent gasket. Accordingly, it is important for conventional fuel cell stacks to be equipped with seals and a suitable compression assembly for applying a compressive force to the seals.

[0007] Conventional methods of sealing around plate manifold openings and MEAs within fuel cells include framing the MEA with a resilient fluid impermeable gas-

EP 1 156 546 A1

15

ket, placing preformed gaskets in channels in the electrode layers and/or separator plates, or molding seals within grooves in the electrode layer or separator plate, circumscribing the electrochemically active area and any fluid manifold openings. Examples of conventional methods are disclosed in U.S. Patent Nos. 5,176,966 and 5,284,718 and European Patent Publication No. 0604683 Al (Application No. 92122145.3). Typically the gasket seals are cut from a sheet of gasket material. For a gasket seal that seals around the electrochemically active area of the MEA, the central portion of the sheet is cut away. This procedure results in a large amount of the gasket material being wasted. Because the electrodes are porous, for the gasket seals to operate effectively, the gasket seals ordinarily are in direct contact with the flow field plates and the ion exchange membrane. Therefore, in a conventional MEA, electrode material is cut away in the sealing regions so that the gasket will contact the ion exchange membrane. Some MEAs use additional thin-film layers to protect the ion exchange membrane where it would otherwise be exposed in the gasket seal areas. Separate components such as gasket seals and thin-film layers requirerespective processing or assembly steps, which add to the complexity and expense of manufacturing fuel cell stacks.

[0008] As an alternative to placing sealant material into grooves, it is known to impregnate a fluid impermeable electrically insulating sealant material into the porous electrode layers of a MEA having its electrode layers coextensive with its ion exchange membrane, see e.g. U.S. 5,523,175. WO 98/33225 Al, published after the priority date of the present application, discloses another MEA of this type with the sealant material extending laterally beyond the membrane and electrode layers, so as to envelope the peripheral region of both electrodes and the membrane. The sealant is applied and cured in a mold. The molded seal may extend axially beyond the electrode surfaces opposite the membrane, to provide spaces for receiving gas distribution structures between the respective electrode and a bipolar plate abutting against the projecting face of the seal.

[0009] EP 0 604 683 A1 teaches a method of making a membrane electrode assembly with resilient Integral seals for use in an electrochemical fuel cell, wherein said membrane electrode assembly is made by bonding a solid polymer ion exchange membrane between two layers of porous electrically conductive sheet material, the layers of sheet material covering and supporting substantially the entire surface of the membrane, and impregnating the surfaces of the layers of sheet material facing away from the membrane with a sealant material, the sealant material generally circumscribing fluid passage openings formed in the layers of sheet material and the membrane to accommodate the passage of fluids through the assembly.

[0010] With respect to the technique of impregnating sealing regions of a preferably porous electrode, it is

known to apply overpressure on the sealant material to facilitate introducing the same and impregnating the electrode, see e.g. U.S. 4,588,661 and WO 92/03854 A2.

5 [0011] Accordingly, it is the technical problem underlying the present invention to provide a novel method of making a membrane electrode assembly for an electrochemical fuel cell, capable to simplify and reduce the number of individual or separate components involved in sealing in a fuel cell stack, since this reduces assembly time and the cost of manufacturing, while achieving a reliable sealing effect.

Summary of the Invention

[0012] The invention solves this problem by providing a method of making a membrane electrode assembly according to claim 1.

[0013] In a preferred embodiment the sealing regions comprise regions that circumscribe the electrochemically active area of the electrode layers.

[0014] If the MEA further comprises one or more openings formed therein, such as opening for a fluid manifold and/or a tension member, the sealing region further may comprises regions that circumscribe such openings.

[0015] In a fuel cell stack the sealing regions cooperate with the fuel cell separator plates to prevent fluids from leaking around the edges of the MEA. The sealant material is preferably an elastomer. In a preferred method of making an improved MEA, the sealant material is injection molded. Accordingly, it is desirable for the uncured sealant material to be flow processable. After the uncured sealant material has been applied to the MEA, it is allowed to cure to form a resilient elastomeric material. The clastomeric sealant material may be a thermosetting material, as long as the curing temperature is compatible with the MEA components, and in particular, the ion exchange membrane.

[0016] The sealant material may also be used to form a reference feature such as a raised edge or protrusion for assisting in the assembly of the fuel cell. For example, when an outer perimeter edge seal is being molded, at least one of the edges could be molded with a reference edge, which can be used to align the MEA during manufacturing processes. Alternatively, a protrusion such as a cylindrical plug could be molded in a location, which can be aligned with a corresponding cylindrical depression in an adjacent separator plate during stack assembly.

[0017] From a manufacturing perspective, the coextensive electrode and membrane of the improved MEA provides advantages for high-speed manufacturing. For example, the electrode and membrane layers of the MEA may be formed in continuous processes that produce a multi-layered roll of material or large sheets that can be cut down to the size of an individual MEA. This is difficult in conventional MEAs where the electrodes

15

35

are not coextensive with the membrane (i.e. because the membrane extends laterally beyond the edge of the electrodes). However, because the electrodes and membrane are coextensive, the improved MEAs may be "flush cut" from a larger piece of multi-layered mate-

5

[0018] An improved MEA with coextensive layers has been successfully manufactured by flush-cutting without short circuiting problems arising. Preferred aspects of the manufacturing process that have contributed to this success include one or more of the following:

- (a) after flush cutting the multi-layer material, brushing the edges of the flush cut material to remove tiny portions of the electrode layer that may extend laterally beyond the edge of the membrane;
- (b) using a vacuum during the impregnation or injection molding process;
- (c) enveloping sealing regions of the MEA with a sealant material; and
- (d) forming the integral seal and then flush cutting in the sealing regions.

[0019] It is believed that employing at least one of these approaches in the method of manufacturing an improved MEA provides significant benefits that help to prevent short circuiting caused by tiny particles of the electrodes straddling the membrane. For example, brushing the edges of the flush cut material may result in the removal of portions of the electrode layer that extend laterally beyond the edge of the membrane. Applying a vacuum to the outer surfaces of the porous electrode layers during the impregnation or injection molding process helps to direct the flow of the sealant material so as to pull the electrode material away from the membrane and the opposing electrode while the sealant material is being applied. Another advantage of applying a vacuum is that it helps to remove air from the mold and reduce the effect of bubble formation or foaming in the sealant material. Furthermore, short circuiting may be reduced by enveloping peripheral regions of the MEA with an electrically insulating sealant material. The sealant material embeds the cut edge of the MEA so that the edges of the electrode layers are electrically insulated. The embedded electrode material is also immobilized by the sealant material so that the edges of the electrodes can not be displaced by fluid currents or pressures within an operating fuel cell.

[0020] An additional benefit of enveloping peripheral regions of the MEA with the integral seal is that the fluid impermeable sealant material prevents dehydration of the membrane through the side edge.

[0021] In preferred embodiments, the integral seal further comprises a raised rib that is compressible when the MEA is placed between opposing fuel cell plates in an assembled fuel cell. The raised rib may cooperate with a recessed groove in the plate that provides a recessed sealing surface. The advantage of using a re-

cessed sealing surface is that it is less susceptible to being damaged because the sealing surface is somewhat protected by being recessed. For example, a recessed sealing surface is less likely to be scratched when fuel cell plates are stacked on top of each other while being fabricated. Scratches in the sealing surface could cause leaks in an operating fuel cell. Ribs may be located in sealing regions where the sealant material is impregnated into the porous electrode layers (i.e. superposing the membrane) and/or may be located in regions where the sealant material extends laterally beyond the edges of the electrode layers and membrane.

[0022] In preferred embodiments, the integral seal of the MEA comprises a plurality of the raised ribs. For example, the ribs in each sealing region may be parallel, with each individual rib circumscribing the active area or the openings in the MEA. An advantage of having a plurality of raised ribs is increased protection against leaks. Each one of the plurality of ribs must be breached for there to be a fluid leak.

[0023] Versions of these preferred embodiments also employ raised cross-ribs between adjacent ones of the plurality of raised ribs. The cross-ribs compartmentalize the spaces between the raised ribs. Therefore, for there to be a leak, there must be a breach in the raised ribs adjoining the same sealed compartment; otherwise, any fluid leaking through a breach in a raised rib will be confined to the sealed compartment.

[0024] A method of making a MEA with resilient inteo gral seals for use in an electrochemical fuel cell comprises the following sequential steps:

- (a) placing the MEA inside a mold;
- (b) introducing a curable flow processable sealant material into the mold;
- (c) directing the sealant material to desired sealing regions of the MEA and impregnating a portion of an electrode layer of the MEA with the sealant material in the sealing regions; and
- (d) curing the sealant material to form an integral seal; and
- (e) removing the MEA from the mold.

[0025] In a preferred method step (c) is accompanied by the step of applying a vacuum to assist the injection and impregnation of the sealant material. If the vacuum is applied to both major surfaces of the MEA, the vacuum also helps to prevent electrode material from bridging between the electrodes during the injection process. [0026] A preferred method comprises using a sealant material that is a thermosetting material. This embodiment of the method further comprises the step of applying heat until the sealant material is cured. To cure the sealant material while it is still within the mold, the temperature must be controlled to prevent overheating that may damage the MEA, and in particular the ion exchange membrane.

[0027] The mold preferably has open channels

formed in the molding surface. The channels facilitate the distribution of the curable flow processable sealant material to the sealing regions. The channels also act as molding surfaces for forming ribs or ridges in the integral seal. The mold is also preferably fitted with a raised dike. The dike impinges upon the MEA to limit the extent to which the sealant material impregnates the electrode layer. For example, the mold may have opposing dikes on opposite surfaces of the mold. When the mold is closed, the dikes press against and compress the electrode layers. The compressed electrode layers have reduced porosity, which helps to confine the impregnation of the electrode layers to the sealing regions. The dikes are thus positioned on the sides of the sealing regions that face the electrochemically active areas of the MEA.

Brief Description Of The Drawings

[0028] The advantages, nature and additional features of the invention will become more apparent from the following description, together with the accompanying drawings, in which:

FIG. 1 is a partially exploded isometric view of an electrochemical solid polymer fuel cell stack that incorporates an embodiment of an improved membrane electrode assembly that has integral fluid impermeable seals;

FIG. 2 is a plan view of an improved membrane electrode assembly;

FIGS. 3A through 3D are partial section views of an edge portion of the membrane electrode assembly of FIG. 2, as indicated by the section marked in FIG. 2.

FIGS. 4A through 4C are partial section views of the edges of three embodiments of a membrane electrode assembly interposed between two fuel cell separator plates with integral seals compressed therebetween; and

FIG. 5 is an enlarged plan view of a portion of a preferred embodiment of a membrane electrode assembly that has an integral seal that incorporates a plurality of seal ridges and cross-ridges.

Detailed Description of the Preferred Embodiments

[0029] FIG. 1 illustrates a solid polymer electrochemical fuel cell stack 10, including a pair of end plate assemblies 20 and 30, and a plurality of stacked fuel cell assemblies 50, each comprising an MEA 100, and a pair of flow field plates 200. A tension member 60 extends between end plate assemblies 20 and 30 to retain and secure stack 10 in its assembled state. Spring 70 with clamping members 80 grip each end of tension member 60 to apply a compressive force to fuel cell assemblies 50 of stack 10.

[0030] Fluid reactant streams are supplied to and ex-

hausted from internal manifolds and passages in stack 10 via inlet and outlet ports 40 in end plate assemblies 20 and 30. Aligned openings 105 and 205 in MEAs 100 and flow field plates 200, respectively, form reactant manifolds extending through stack 10.

[0031] In the illustrated embodiment, an integral perimeter seal 110 is provided around the outer edge of MEA 100. Integral manifold seals 120 circumscribe manifold openings 105. When stack 10 is secured in its assembled, compressed state, integral seals 110 and 120 cooperate with the adjacent pair of plates 200 to fluidly isolate fuel and oxidant reactant streams in internal reactant manifolds and passages, thereby isolating one reactant stream from the other and preventing the reactant streams from leaking from stack 10.

[0032] As illustrated in FIG. 1, each MEA 100 is positioned between the active surfaces of two flow field plates 200. Each flow field plate 200 has flow field channels 210 on the active surface thereof (which contacts the MEA) for distributing fuel or oxidant fluid streams to the active area of the MEA 100. In the embodiment illustrated in FIG. 1, flow field channels 210 are fluidly connected to manifold openings 205 in plate 200 via supply/exhaust channels 220 (partially shown) located on the non-active surface of flow field plate 200 and ports 230 extending through plate 200.

[0033] In the illustrated embodiment, flow field plates 200 have a plurality of open-faced parallel channels 250 formed in the non-active surface thereof. Channels 250 on adjacent pairs of plates 200 cooperate to form passages extending through stack 10, through which a coolant stream, such as air, may be directed.

[0034] FIG. 2 shows an MEA 100 with integral seals 110, 120 that respectively circumscribe the electrochemically active area of MEA 100, and manifold openings 105 and opening 115 through which tension member 60 extends. MEA 100 comprises an ion exchange membrane (not visible in FIG. 2) disposed between two porous, electrically conductive electrode layers 140. These electrode layers 140 may, for example be carbon fiber paper. A sealant material, preferably a flow processable elastomer, such as, for example, a thermosetting liquid injection moldable compound (e.g. silicones, fluoroelastomers, fluorosilicones, ethylene propylene di-methyl, and natural rubber), is impregnated into the porous electrode layers of MEA 100 to form integral seals 110 and 120.

[0035] Various embodiments of an MEA 100 with an integral seal such as 110, are illustrated in cross-sectional views in FIGS. 3A through 3D. The figures depict a perimeter edge integral seal 110, such as through section 3-3 of FIG. 2, although the same configurations could also be employed for integral seal 120 at a manifold opening (as in FIG. 1). Each embodiment of an MEA 100 comprises an ion exchange membrane 130 disposed between two porous, electrically conductive electrode layers 140, and a sealant material 125 impregnated into a portion 150 of the porous electrode layers

of MEA 100. Preferably, at least a portion of seal 110 protrudes above the outer surface of porous electrode layers 140.

9

[0036] In all of the illustrated embodiments 3A through 3D, porous electrode layers 140 extend to the edge of ion exchange membrane 130. That is, the electrode layers 140 and the ion exchange membrane 130 are coextensive. The multi-layer MEA 100 may be assembled and then cut to the desired shape and dimensions; then the sealant material 125 may be impregnated into a portion 150 of the porous electrode layers 140. Alternatively, sealant material 125 can be impregnated into a sheet of MEA material. The integral seals for a plurality of MEAs could be injection molded onto the sheet of MEA material, impregnating a plurality of sealing regions of the porous electrode layers 140. After sealant material 125 has cured, the MEA 100 and sealant material 125 may both be cut (preferably in the sealing regions) to the desired dimensions at the same time. Because the sealant material was injection molded prior to the ion exchange membrane being cut, the two electrode layers are kept apart while the sealant material is being injected. Thus the electrode material in the sealing regions is embedded within the electrically insulating sealant material. Cutting the multi-layer material in the sealing regions after the sealant material cures, helps to prevent the possibility of short-circuiting because the cured sealant material immobilizes the embedded electrode material.

[0037] In the embodiment of FIG. 3A, integral seal 110 extends only as far as the edge of ion exchange membrane 130. That is, the edge of seal 110 is flush with the edge of membrane 130 and electrode layers 140. Therefore, the embodiment shown in FIG. 3A may be made by applying the sealant material before or after the MEA 100 is cut to the desired size and shape.

[0038] FIG. 3B illustrates a preferred embodiment. Similar to the embodiment of FIG. 3A, ion exchange membrane 130 is coextensive with porous electrode layers 140 and sealant material is impregnated into a portion 150 of porous electrode layers 140. Unlike the embodiment of FIG. 3A, sealant material 125 extends laterally beyond the edge of MEA 100, enveloping the edge of ion exchange membrane 130. By enveloping the edge, the sealant material 125 contacts three surfaces of ion exchange membrane 130, namely portions of the two surfaces that face the two electrodes 140 and the side edge defined by the thickness of membrane 130. Integral seal 110 has a single raised rib 160, in the region of the seal that extends beyond the membrane. FIG. 3B also shows an alignment feature in the form of a cylindrical plug or pin 162. Sealant material may be used to make plug 162 that may be molded and formed at the same time as integral seal 110. Plug 162 can cooperate with a corresponding cylindrical depression or well in the adjacent separator plate of a fuel cell to facilitate alignment of MEA 100 with the separator plates during assembly of the fuel cell.

[0039] FIG. 3C illustrates an embodiment of an integral seal 110 that has some of the same features as the embodiment depicted by FIG. 3B. However, instead of having only one rib on each face, the embodiment of FIG. 3C has three spaced ribs 165, 170, 175 and crossribs 180. Those skilled in the art will appreciate that additional ribs will increase the protection against leaks. A breach in one of the ribs will not result in a leak unless there are also breaches in the other parallel ribs. The benefit of the plurality of ribs is augmented by the crossribs 180 that compartmentalize the spaces between parallel ribs 165, 170, 175. With the compartmentalized spaces, a leak will not occur unless there is a breach in all three of ribs 165, 170, and 175 within the same compartment between a pair of spaced cross-ribs 180.

[0040] FIG. 3D illustrates a preferred embodiment of an integral seal 110 that shares some of the same features as the embodiments depicted by FIGS. 3B and 3C. However, one of the pairs of raised ribs is located in the sealing region that overlaps an edge portion of electrode layers 140 and superposes the membrane. An advantage of this embodiment is that the mechanical pressure that compresses ribs 190 exerts a pinching force on membrane layer 130 to help prevent fluid leaks around the edge of membrane layer 130. The embodiment of FIG. 3D may also employ cross-ribs 195 for further protection against fluid leaks. FIG. 3D also illustrates the feature of a raised reference edge 197, which may be formed from the sealant material. Reference edge 197 may be used to assist with aligning the MEA with the adjacent fuel cell components, which may be shaped to engage with reference edge 197. Alternatively, reference edge 197 may be used during the manufacturing process to seat the MEA against a guide surface of a machine used to assemble the fuel cells.

[0041] FIGS. 4A through 4C show an MEA 100 with an integral seal compressed between two fuel cell separator plates 200. Because at least a portion of MEA integral seal 110 is thicker and/or firmer than MEA 100, the compressive forces acting on the fuel cell stack compress seal 110 against active surfaces 260 of separator plates 200. FIG. 4A depicts the MEA of FIG. 3A compressed between two separator plates 200. FIG. 4B depicts the MEA of FIG. 3B compressed between two separator plates 200. FIG. 4B illustrates an embodiment of the invention wherein surface 260 of plate 200 includes a recessed groove 265. An advantage of this arrangement is that the recessed surface is less prone to scoring or other damage that may occur during the manufacturing process when a number of flow field plates 200 may be stacked one on top of the other. FIG. 4C depicts the MEA of FIG. 3C compressed between two flow field plates 200. FIG. 4C shows that the spacing between parallel ribs 165, 170, and 175 is sufficient to accommodate lateral bulging of the ribs under compression and still provide spaces therebetween.

[0042] FIG. 5 is a partial plan view of an MEA 100 with an integral perimeter edge seal 110 such as that shown

35

40

in section view by FIG. 3C. Three spaced parallel ribs 165, 170 and 175 circumscribe the active area of MEA 100. Spaced cross-ribs 180 provide fluidly isolated compartments 185 between ribs 165, 170 and 175.

11

[0043] The scope of the invention is to be construed in accordance with the substance defined by the following claims.

8. The method of claim 1 wherein said sealant material is a thermosetting material and said method further comprises the step of applying heat to cure said thermosetting material.

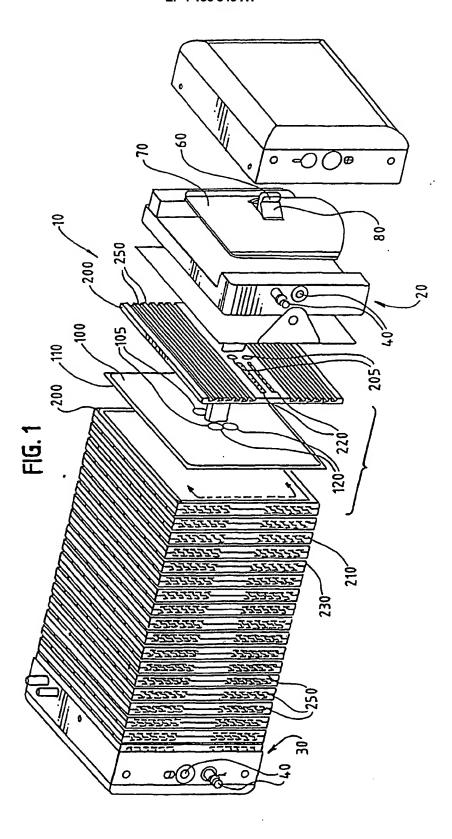
Claims

A method of making a membrane electrode assembly with resilient integral seals for use in an electrochemical fuel cell, wherein said membrane electrode assembly is made from a multi-layer material comprising an ion exchange membrane interposed between two electrode layers and an electrocatalyst disposed on interfacing surfaces between said membrane and said first and second electrode layers; said method comprising the following sequential steps:

20

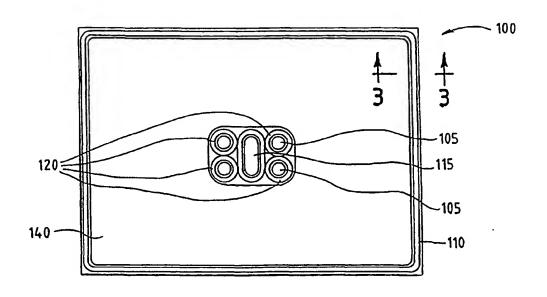
(a) impregnating sealing regions of said first and second electrodes with a fluid impermeable curable flow processable sealant material, while applying a vacuum to facilitate introducing said sealant material and impregnating said first and second electrodes;

- (b) curing said sealant material; and
- (c) cutting through said multi-layer material in 30 said sealing regions.
- The method of claim 1 wherein said sealing regions define a size and shape of said membrane electrode assembly.
- The method of claim 2 wherein said sealing regions further define the size and shape of openings in said membrane electrode assembly.
- The method of claim 1 wherein said sealant material is injected into a mold to impregnate said electrode layers of said multi-layer material.
- The method of claim 3 wherein said mold provides
 channels for forming sealing ribs that protrude
 above the surface of said electrode layers.
- The method of claim 3 further comprising the step of applying a vacuum to the interior of the mold to assist with injecting said sealant material into said mold.
- The method of claim 4 further comprising employing a raised dike in said mold wherein said dike impinges upon said multi-layer material to limit the extent to which said sealant material impregnates said electrode layer.

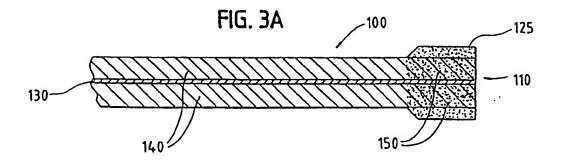


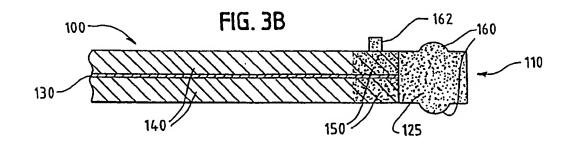
つ

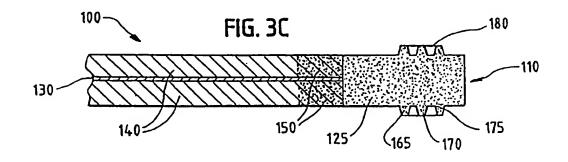
FIG. 2

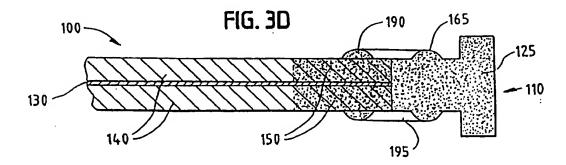


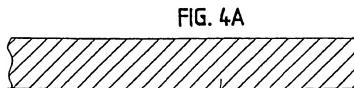
フ

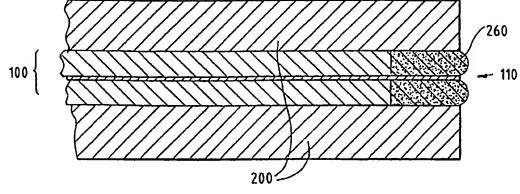


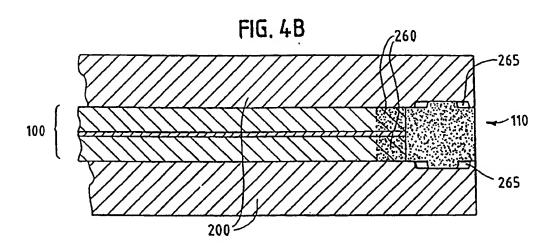












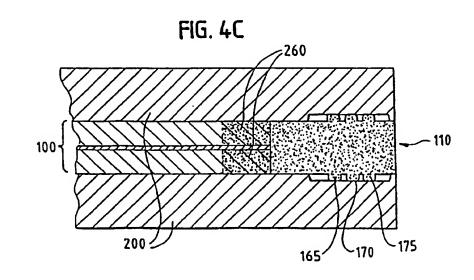
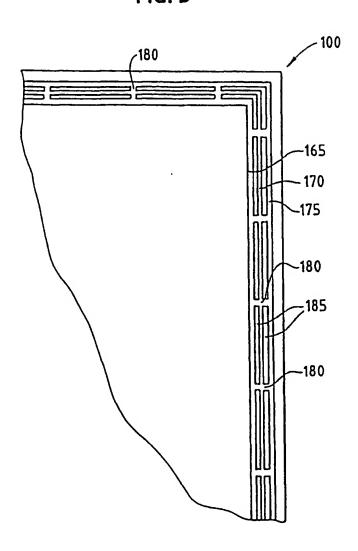


FIG. 5





EUROPEAN SEARCH REPORT

Application Number EP 01 11 9398

		ERED TO BE RELEVANT	r	
Category	Citation of document with is of relevant pass	ndication, where appropriate, ages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.CI.7)
Y	US 5 523 175 A (BEA 4 June 1996 (1996-0 * column 3, line 63	_	1-8	H01M8/24 H01M8/02 H01M8/10
Y	EP 0 604 683 A (BAL 6 July 1994 (1994-0 * column 4, line 29 * column 5, line 9- * column 5, line 51	7-06) -42 *	1-8	
A	US 5 219 674 A (SIN 15 June 1993 (1993- * column 2, line 41		1-8	
E	WO 98 33225 A (MAGN ARTHUR (DE); SCHWES 30 July 1998 (1998- * page 14, line 1-1 * page 15, line 3 -	07-30) 4 *	1-8	
A	WO 92 03854 A (INT 5 March 1992 (1992- * claims; figures * * page 21, line 22	03-05)	1-8	TECHNICAL FIELDS SEARCHED (Int.CI.7) H01M
A	13 May 1986 (1986-0	FMAN ARTHUR ET AL) 5-13) - column 3, line 5 *	1-8	
	The present search report has	been drawn up for all claims		
	Place of search	Date of completion of the search		Examiner
	MUNICH	25 September 2003	l Eng	11, H
X : part Y : part doc A : tech O : nor	ATEGORY OF CITED DOCUMENTS doularly relevant it taken alone localarly relevant it combined with anot ument of the same category wological background inwritten disclosure mediate document	T : theory or principle E : earlier patent doc after the filling dat ber D : document etecl is L : document afted for 8 : member of the si document	ament, but publi the application or other reasons	Ished on, or

PO FORM 1509 09.82 (P

ANNEX TO THE EUROPEAN SEARCH REPORT ON EUROPEAN PATENT APPLICATION NO.

EP 01 11 9398

This armsx itsis the patent family members relating to the patent documents cited in the above—mentioned European search report. The members are as contained in the European Patent Office EDP file on The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

25-09-2001

Patent document cited in search report		Publication date	Publication date		Patent family member(s)		
US	5523175	Α	04-06-1996	WO	9313566	A1	08-07-1993
EP	0604683	A	06-07-1994	ΕP	0604683	A1	06-07-1994
				ΑT	180105	T	15-05-1999
				DE	69229177	D1	17-06-1999
				DE	69229177	T2	02-12-1999
US	5219674	A	15-06-1993	CA	2126681	A1	08-07-1993
				EΡ	0619045	A1	12-10-1994
				JΡ	7502374		09-03-1995
				₩O	9313567	A1	08-07-1993
WO	9833225	A	30-07-1998	DE	19703214		05-11-1998
				AU	6617298		18-08-1998
				CN	1251212	T	19-04-2000
				WO	9833225		30-07-1998
				EP	0966770		29-12-1999
				JP	2001509304	T	10-07-2001
WO	9203854	A	05-03-1992	CA	2090477		01-03-1992
				EP	0546116		16-06-1993
				MO	9203854	A2 	05-03-1992
US	4588661	A	13-05-1986	AT	42652	•	15-05-1989
				CA	1250017		14-02-1989
				DE	3569852		01-06-1989
				EΡ	0174762		19-03-1986
				JP	1939090	-	09-06-1995
				JP	6065049		22-08-1994
				JP	61061372	A 	29-03-1986
	•						
						•	

For more details about this annex : see Official Journal of the European Patent Office, No. 12/82

This Page is Inserted by IFW Indexing and Scanning Operations and is not part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

□ BLACK BORDERS
□ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
□ FADED TEXT OR DRAWING
□ BLURRED OR ILLEGIBLE TEXT OR DRAWING
□ SKEWED/SLANTED IMAGES
□ COLOR OR BLACK AND WHITE PHOTOGRAPHS
□ GRAY SCALE DOCUMENTS
□ LINES OR MARKS ON ORIGINAL DOCUMENT
□ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY

IMAGES ARE BEST AVAILABLE COPY.

☐ OTHER:

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.